

C. Amendment to the Specification

Please amend the paragraphs on page 1, line 18 - page 2, line 14, as follows:

An old example of organic luminescence device is, e.g., one using luminescence of a vacuum-deposited anthracene film (Thin Solid Films, 94 (1982) 171). In recent years, however, in view of the advantages[[],] such as easiness of providing a large-area device compared with an inorganic luminescence device[[],] and a possibility of obtaining realizing desired luminescence colors in view of the by development of various new materials and drivability at low voltages, an extensive study related to forming thereon for device formation as a luminescence device of a high-speed responsiveness and a high efficiency[[],] has been conducted.

As described in detail in, e.g., Macromol. Symp. 125, 1-48 (1997), an organic EL device generally has a structure comprising upper and lower two electrodes and a plurality of organic film layers between the electrodes formed on a transparent substrate. Basic structures thereof are shown in Figures 1(a) and (b).

As shown in Figure 1, an organic EL device generally has a structure comprising a transparent[[],] electrode 14, a metal electrode 11, and a plurality of organic film layers therebetween on a transparent substrate 15.

Please amend the paragraph on page 2, line 26 - page 3, line 6, as follows:

For the luminescence layer 12, aluminum quinolynol quinolynol complexes (a representative example thereof is Alq3 shown hereinafter), etc., having an electron-transporting characteristic and luminescence characteristic are used. For the hole-transporting layer, biphenyldiamine derivatives (a representative example thereof is .alpha.-NPD shown hereinafter), etc., having an electron-donative characteristic are used.

Please amend the paragraphs on page 4, lines 3-17, as follows:

Known luminescence processes used heretofore in organic EL devices include one utilizing an excited singlet state and one utilizing an excited triplet state, and the transition from the former state to the ground state is called "fluorescence" and the transition from the latter state to the ground state is called "phosphorescence". And the substances in these excited states are called a singlet exciton and a triplet exciton, respectively.

In most of the organic luminescence devices studied heretofore, fluorescence caused by the transition from the excited singlet state to the ground state[[],] has been utilized. On the other hand, in recent years, devices utilizing phosphorescence via triplet excitons have been studied.

Please amend the paragraphs on page 6, line 6 - page 7, line 4, as follows:

The above-mentioned Articles 1 and 2 both have reported structures[[],] as exhibiting a high efficiency, including a hole-transporting layer 13 comprising α -NPD, an electron-transporting layer 16 comprising Alq3, an exciton diffusion-preventing layer 17 comprising BCP, and a luminescence layer 12 comprising CBP as a host and ca. 6% of

platinum-octaethylporphyrin complex (PtOEP) or iridium-phenylpyrimidine complex (Ir(ppy)₃) as a phosphorescent material dispersed in mixture therein.

Such a phosphorescent material is particularly noted at present because it is expected to provide a high luminescence efficiency in principle for the following reasons. More specifically, excitons formed by a carrier recombination comprise singlet excitons and triplet excitons in a probability ratio of 1:3. Conventional organic EL devices have utilized fluorescence of which the luminescence efficiency is limited to at most 25%. On the other hand[[,]] if phosphorescence generated from triplet excitons is utilized, an efficiency of at least three times is expected, and even an efficiency of 100%, i.e., four times, can be expected in principle, if a transition owing to the intersystem crossing from a singlet state having a higher energy to a triplet state is taken into account.

Please amend the paragraph on page 7, lines 10-12, as follows:

The reason for of the deterioration has not been fully clarified, but the present inventors consider it to be as follows based on the mechanism of phosphorescence.

Please amend the paragraphs on page 8, lines 4-21, as follows:

Particularly, in a phosphorescent material, this may be attributable to a life of the triplet excitons, which is longer by three or more digits than the life of a singlet exciton. More specifically, because it is held in a high-energy excited state for a longer period, it is likely liable to react with surrounding materials and cause polymer formation

among the excitons, thus incurring a higher probability of a deactivation process resulting in a material change or life deterioration, as we have considered.

Needless to say, a luminescence efficiency of an organic luminescence device is increased by increasing the luminescence quantum yield of a luminescence center material, but it is also an important factor for enhancing the luminescence intensity of the device to increase the concentration of a luminescence material in the luminescence layer.

Please amend the paragraph on page 9, line 15 - page 10, line 3, as follows:

The phenomenon is caused by abundant presence of molecules in the triplet excited state waiting for luminescence in the case of a phosphorescence substance having a life of triplet exciton longer by 3 digits or more than the life of the singlet exciton. In this state, thermal deactivation of losing energy due to a mutual interaction of triplet excitons is likely liable to occur. This is called triplet-triplet extinction and is associated with a lowering in luminescence efficiency at a high current density. Further, it is also considered that due to a long retention time at a high energy state, the excitons ~~are caused to~~ have an increased probability of reacting reaction with a surrounding material and forming formation of polymers of excitons, thereby causing deactivation, or even leading to a material change or a deterioration of life.

Please amend the paragraph on page 10, lines 12-18, as follows:

More specifically, an object of the present invention is to provide a luminescence material less likely to lead liable to cause concentration extinction even when used at a high concentration with respect to the host material in a luminescence layer by

introducing a substituent group into a metal coordination compound as such a luminescence material.

Please amend the paragraphs on page 12, line 14 - page 13, line 8, as follows:

According to another aspect, the organic luminescence device of the present invention is a luminescence device comprising at least one layer or plural layers of organic film disposed between a cathode and an anode and including at least one ~~layer of~~ luminescence layer, that is characterized by containing a luminescence molecule of the formula (1) having a substituent and showing a maximum luminescence characteristic at a concentration higher than a concentration at which a luminescence molecule of a similar structure but having no substituent shows a maximum luminescence efficiency.

More specifically, in a luminescence device comprising at least one or plural layers of organic film layer between a cathode and an anode, including at least one ~~layer~~ of luminescence layer, it is preferred that a luminescence molecule of the formula (1) including at least one cyclic group having a substituent is contained at a concentration higher than a concentration at which a luminescence molecule of a similar structure shows a maximum luminescence efficiency.

Please amend the paragraph on page 15, lines 13-22, as follows:

Further, in the structure of Figure 1(b), an electron-transporting layer 16 is disposed between the metal electrode 11 and the luminescence layer 12 in Figure 1(a). As a result, the luminescence function is separated from the functions of election

transportation and hole transportation to provide a structure exhibiting more effective carrier blocking, thus increasing the luminescence efficiency. The electron-transporting layer 16[[],] may comprise, e.g., an oxadiazole derivative.

Please amend the paragraphs on page 16, lines 6-20, as follows:

The present inventors have discovered got a knowledge that the use of a metal coordination compound including a substituted cyclic group and represented by the above-mentioned formula (1) results in allows a high efficiency luminescence and a lesser likelihood provides less liability of concentration extinction even at a high concentration than the conventional level due to suppression of an inter-molecular interaction.

It has been also found that the suppression of concentration extinction is an effect attributable to a substituent on possessed by the metal coordination compound, and the concentration extinction becomes less likely liable to occur not regardless of the coordination number of ligands but owing to the presence of a substituent on at least one ligand.

Please amend the paragraph on page 20, lines 5-27, as follows:

In the case of a ligand of the present invention having a cyclic group having one or plural fluorine atoms, it becomes possible to shift the luminescence wavelength to a shorter side or a longer side because of a change in the energy gap. If it is assumed for convenience that HOMO/LUMO of metal electron orbits and HOMO/LUMO of ligand electron orbits can be considered separately, it is understood that HOMO/LUMO MOHO/LUMO energy levels of ligand electron orbits are changed by fluorine atoms

having a large electro-negativity to change the energy gap between the HOMO level of the metal and the LUMO level of the ligand, thereby shifting the luminescence from the MCLT state as the lowest excited state to a shorter wavelength side or a longer wavelength side. Accordingly, while a luminescence material exhibiting a stably high quantum yield over a broad wavelength range (blue to red) has not been found, it can be realized by a luminescence material of the present invention, thus being able to provide a luminescence material, which shows a high efficiency at a desired emission wavelength over a board wavelength range (from blue to red).

Please amend the paragraphs on page 21, line 10 - page 22, line 24, as follows:

Further, in the case of using a ligand containing a plurality of fluorine atoms or polyfluoroalkyl groups as substituents, it is considered that a direct interaction between luminescence molecules is suppressed due to electrical repulsion caused by their electrical effect or due to steric hindrance, thereby preventing energy deactivation and concentration extinction.

Further, from the viewpoint of device preparation, a luminescence material having a substituent, particularly a fluorinated substituent, allows easier vacuum deposition due to a lowering in sublimation temperature decrease in the film formation by vacuum deposition, thus providing a great advantage also in this respect.

As a result, as shown in the Examples described hereinafter, by using a luminescence material having a substituent according to the present invention, a stable luminescence for many long hours with suppressed concentration extinction can be

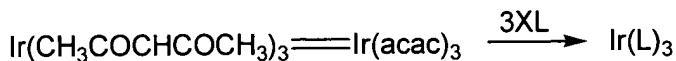
expected. Further, it becomes possible to attain a high phosphorescence yield over a temperature range of from -20°C to 60°C as an actual operation temperature range of an organic luminescence device. Further, in the case of using a compound at a concentration of 8 wt. % or higher with respect to a host material in the luminescence layer or at a concentration higher than a compound having no substituent, it becomes possible to provide an EL device exhibiting excellent luminescence performance while suppressing the concentration extinction. The concentration of the luminescence material of the present invention in the luminescence layer may be at least 8 wt. %, preferably 10 wt. % or higher, but the luminescence material potentially has a possibility of being used even at 100% without causing substantial concentration extinction.

Herein, the term "luminescence performance" refers to a characteristic attributable to a maximum luminescence efficiency that which can be expressed as any of a maximum luminance, a maximum of luminance/current, a maximum of light flux/power consumption or a maximum of external quantum yield.

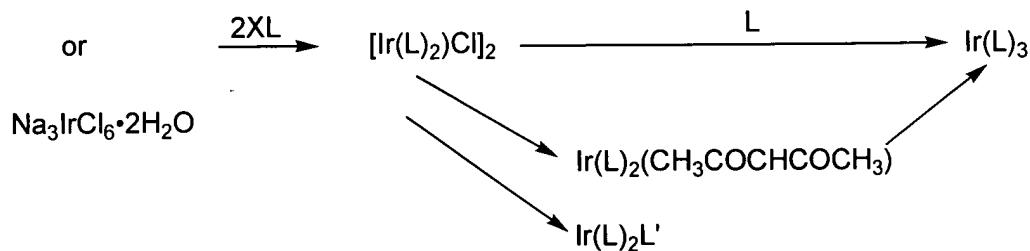
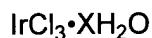
Please amend the paragraph on page 23, lines 18-25, as follows:

For the application to a display, a drive system using a thin-film transistor (abbreviated as TFT) drive circuit according to an active matrix scheme ~~matrix-scheme~~, may be used. By driving a display panel using a luminescence material of the present invention in a luminescence layer, it becomes possible to allow a stable display for many ~~long~~ hours with ~~at~~ a good picture quality.

Please replace the process scheme on page 43, lines 13-22, with the following:



or



Please amend the paragraph on page 52, lines 1-4, as follows:

In order to remove factors for device deterioration due to oxygen or water, the above measurement above-measurement was performed in a dry nitrogen flow after taking the device out of the vacuum chamber.

Please amend the paragraph on page 64, lines 23-26, as follows:

In order to remove factors for device deterioration due to oxygen or water, the above measurement above-measurement was performed in a dry nitrogen flow after taking the device out of the vacuum chamber.

Please amend the paragraph on page 65, line 26, to page 66, line 15, as follows:

In a 200 ml-three-necked flask, 3.50 g (25.0 mmol) of 4-fluorophenylboronic acid, 3.95 g (25.0 mmol) of 1-bromopyridine, 25 ml of toluene, 12.5 ml of ethanol and 25 ml of 2M-sodium carbonate aqueous solution, were placed and stirred at room temperature under a nitrogen stream, followed by addition of 0.98 g (0.85 mmol) of tetrakis(triphenylphosphine)palladium (0). Thereafter, the system was refluxed under stirring and nitrogen stream for 8 hours. After completion of the reaction, the reaction product was cooled and extracted by adding cold water and toluene. The organic layer was washed with saline water and dried on magnesium sulfate, followed by removal of the solvent under a reduced pressure to provide a dry solid. The residue was purified by silica gel column chromatography (eluent: chloroform/methanol=10/1) to obtain 3.24 g (yield=75%) of Compound G.